

Supporting Information for

**Hollow N-doped carbon nanofibers bring superior potassium-storage
performance**

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Supplementary Note

Electrochemical performances of HNCNFs-700 in sodium-ion batteries (SIBs)

Electrochemical measurements

The electrochemical Na-storage performances of HNCNFs-700 were also investigated using coin-type (CR2025) half-cell configurations *vs.* Na metal as the counter electrode, which were assembled in an Ar-filled glove box ($[O_2] < 1$ ppm, $[H_2O] < 1$ ppm). The separator was Whatman glass fiber (GF/D) and the electrolyte was the mixture of ethylene carbonate and diethyl carbonate with a volume ratio of 1:1, containing 1 M $NaClO_4$ and 5 wt% fluoroethylene carbonate. The working electrodes were fabricated by mixing 70 wt% HNCNFs-700, 20 wt% conductive agent (Super P) and 10 wt% binder (Na-CMC) using deionized water as the solvent. The obtained slurry was pasted uniformly on Cu foil and dried in vacuum at 70 °C for 12 h. Then the electrodes were cut into discoidal pieces with a diameter of 12 mm and the mass loading of active materials is 0.4-0.6 mg cm^{-2} . Galvanostatic charge/discharge cycling tests were performed using a LAND CT2001A battery testing system in the voltage range of 0.01-3 V (*vs.* Na^+/Na). Cyclic voltammetry (CV) measurements were carried out on an Ivium-n-Stat electrochemical workstation (Ivium Technologies) with a potential scan rate of 0.1 $mV s^{-1}$ between 0.01-3.0 V (*vs.* Na^+/Na).

Electrochemical performances

The electrochemical performances of HNCNFs-700 as an anode in SIBs are shown in Fig. S14. CV curves of the HNCNFs-700 electrodes were measured at a

scan rate of 0.1 mV s^{-1} within $0.01\text{-}3.0 \text{ V}$ (vs. Na^+/Na) (see Fig. S14a). The broad reduction peak at approximately 0.5 V in the first scan, which disappears at the subsequent scans, can be ascribed to the formation of SEI films. The sharp reversible cathodic peak close to 0.01 V as well as the wide anodic peak are related to the intercalation and deintercalation of Na^+ into/from carbon matrix, respectively. During the following cycles, two broad peaks around 1.2 V and 0.8 V are attributed to the interaction of Na^+ into species of N atoms. The CV curves from the second cycle to the fifth cycle are well overlapped, which demonstrates an excellent reversibility of the HNCNFs-700 electrode. Fig. S14b shows the galvanostatic charge/discharge voltage profiles of the HNCNFs-700 electrode for the 1st, 2nd, 50th, 100th and 200th cycles at 0.1 A g^{-1} . The initial discharge and charge capacities are 788.6 and 345.4 mAh g^{-1} , respectively, corresponding to an initial CE of 43.8% . The HNCNFs-700 electrode delivers a reversible capacity of 308.3 mAh g^{-1} after 200 cycles with CE close to 100% (see Fig. S14c). The HNCNFs-700 electrode also shows superior rate performance with reversible capacities of 323.7 , 278.6 , 245.9 , 229.8 , 218.2 , 209.6 , 188.3 and 169.5 mAh g^{-1} at 0.1 , 0.2 , 0.4 , 0.6 , 0.8 , 1 , 2 and 5 A g^{-1} , respectively (see Fig. S14d). When the current density switches back to 0.1 A g^{-1} , the HNCNFs-700 electrode recovers a specific capacity of 300.8 mAh g^{-1} rapidly and then remains stable for further cycling. Besides, the HNCNFs-700 electrode maintains a high reversible capacity of 223.2 mAh g^{-1} at 1 A g^{-1} after 5000 cycles with CE close to 100% (see Fig. S14e). This suggests HNCNFs-700 is also an excellent anode material for SIBs.

The Na-storage behavior of the HNCNFs-700 electrode was also analyzed by CV measurements at different scan rates from 0.2 to 1 mV s⁻¹ (see Fig. S15a). The calculated *b* values for cathodic and anodic peaks are also both much closer to 1, indicating the Na-storage behavior is dominated by surface capacitive effect. The pseudocapacitive contributions of the HNCNFs-700 electrode at different scan rates are displayed in Fig. S15d.

Supplementary Figures

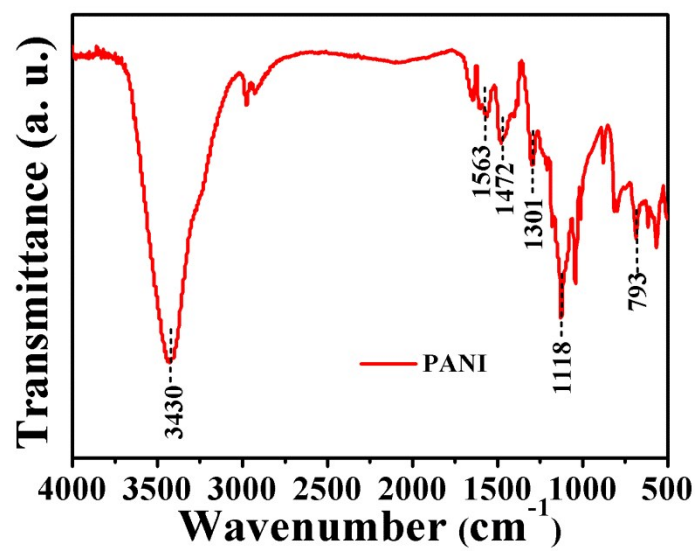


Fig. S1 FT-IR spectrum of PANI nanofibers.

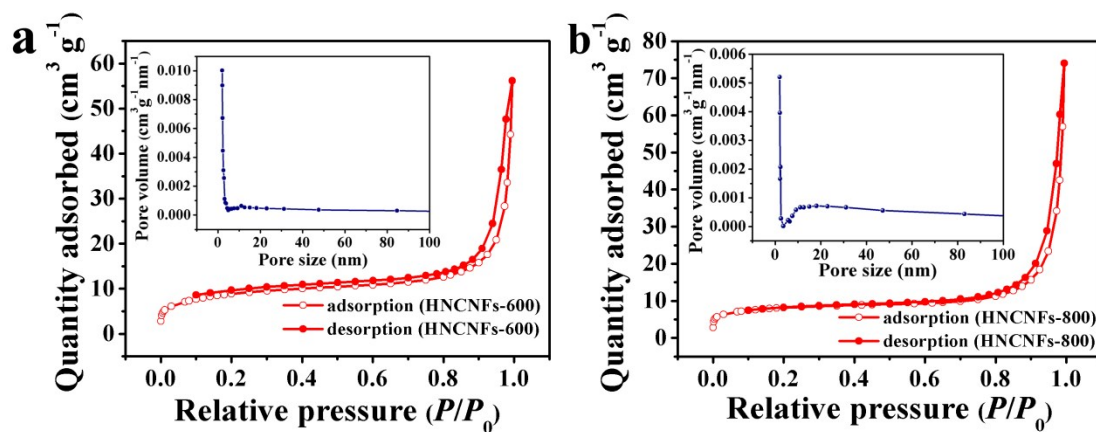


Fig. S2 N₂ adsorption/desorption isotherms and the corresponding pore size distribution (the inset) of (a) HNCNFs-600 and (b) HNCNFs-800.

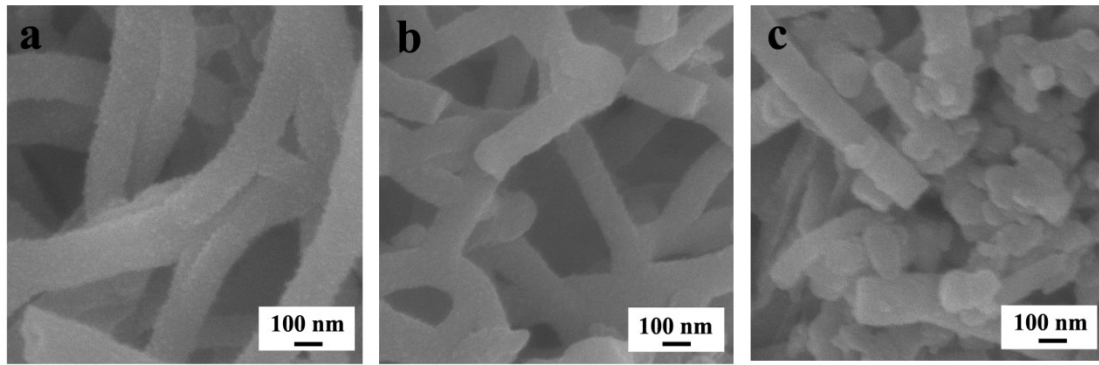


Fig. S3 SEM images of (a) PANI precursor, (b) HNCNFs-600, and (c) HNCNFs-800.

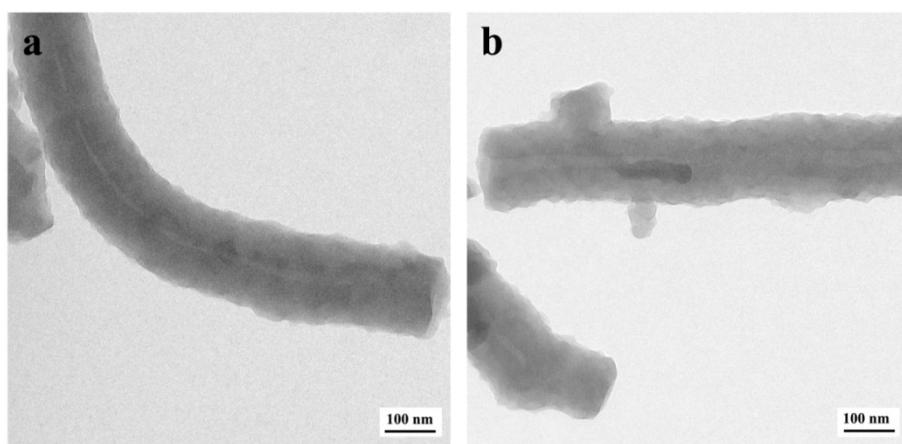


Fig. S4 TEM images of (a) HNCNFs-600 and (b) HNCNFs-800.

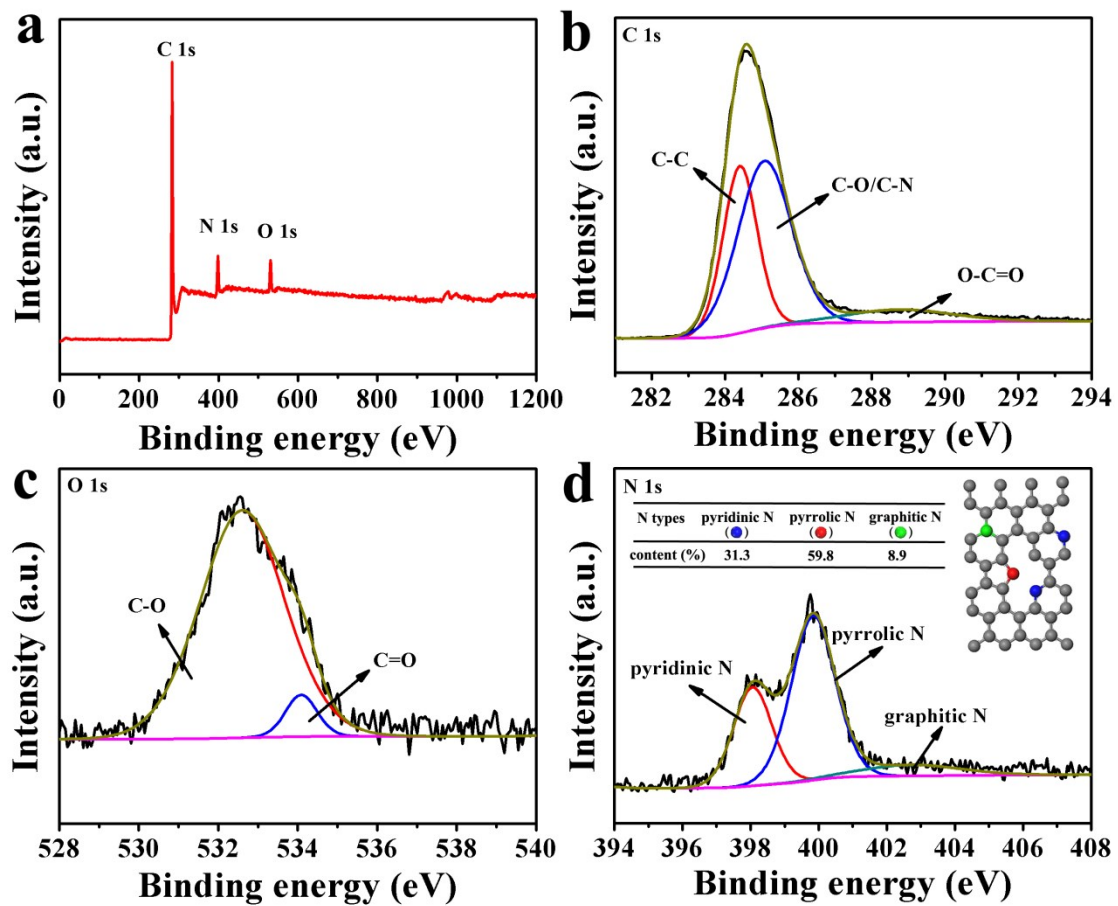


Fig. S5 (a) XPS survey spectrum of HNCNFs-600. (b), (c) and (d) are high-resolution XPS spectra of C 1s, O 1s and N 1s, respectively.

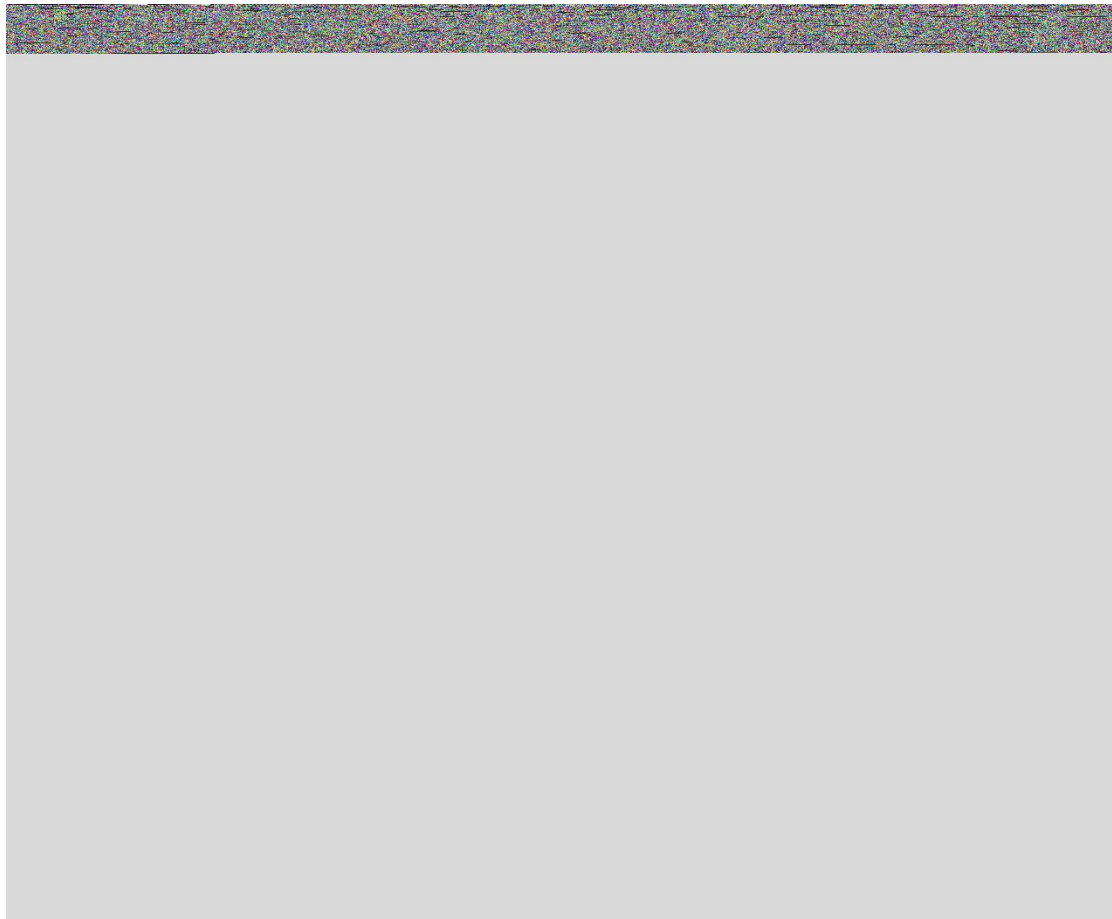


Fig. S6 (a) XPS survey spectrum of HNCNFs-800. (b), (c) and (d) are high-resolution XPS spectra of C 1s, O 1s and N 1s, respectively.

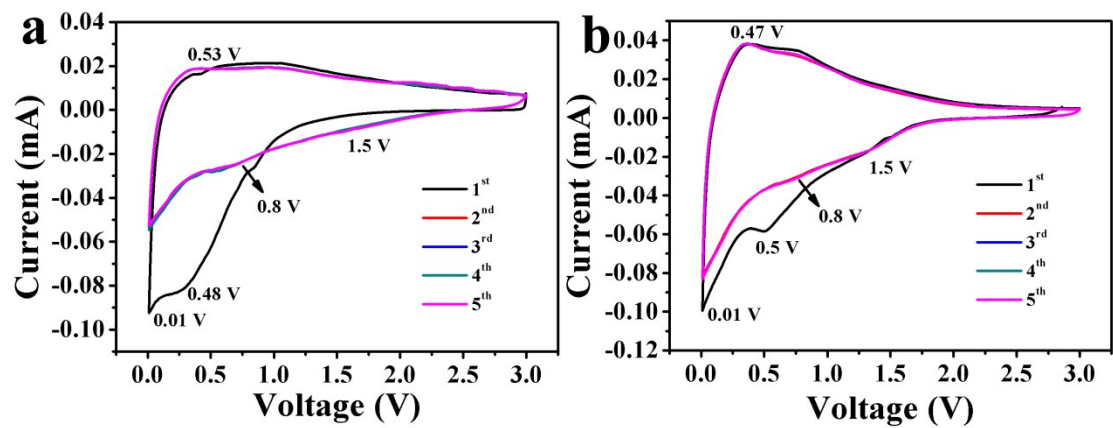


Fig. S7 CV curves of (a) HNCNFs-600 and (b) HNCNFs-800 electrodes at a scan rate of 0.1 mV s⁻¹.

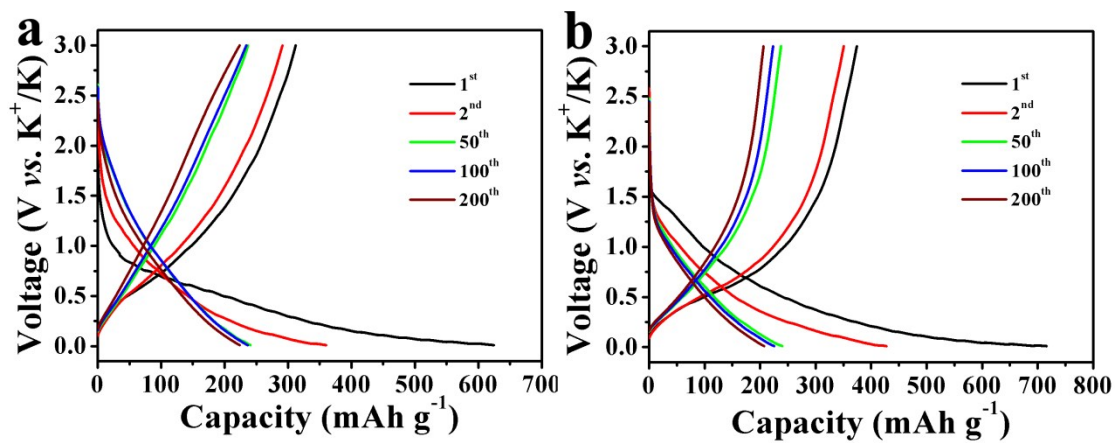


Fig. S8 Galvanostatic charge/discharge curves of (a) HNCNFs-600 and (b) HNCNFs-800 electrodes for the 1st, 2nd, 50th, 100th and 200th cycles at a current density of 0.1 A g⁻¹.

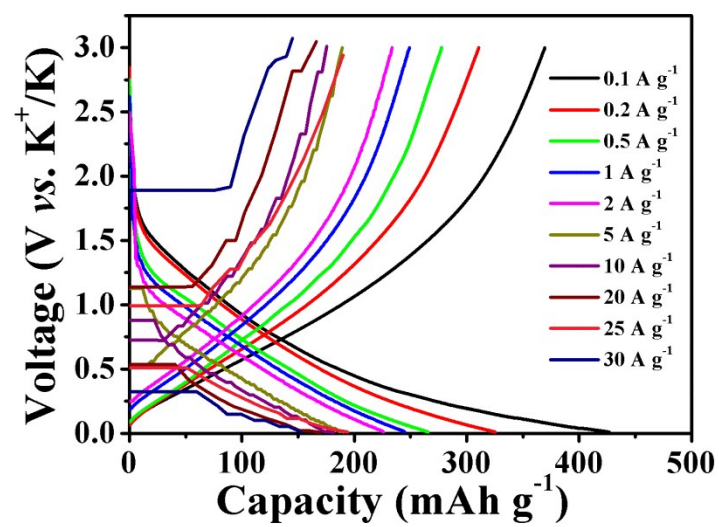


Fig. S9 Galvanostatic charge/discharge curves of the HNCNFs-700 electrode at various current densities.

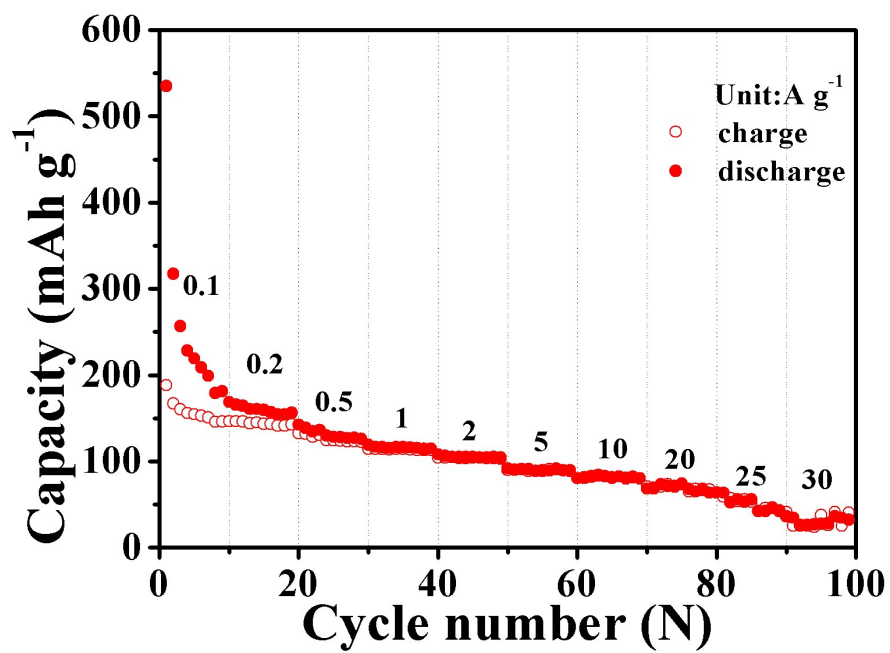


Fig. S10 Rate performance of the super P electrode at various current densities.

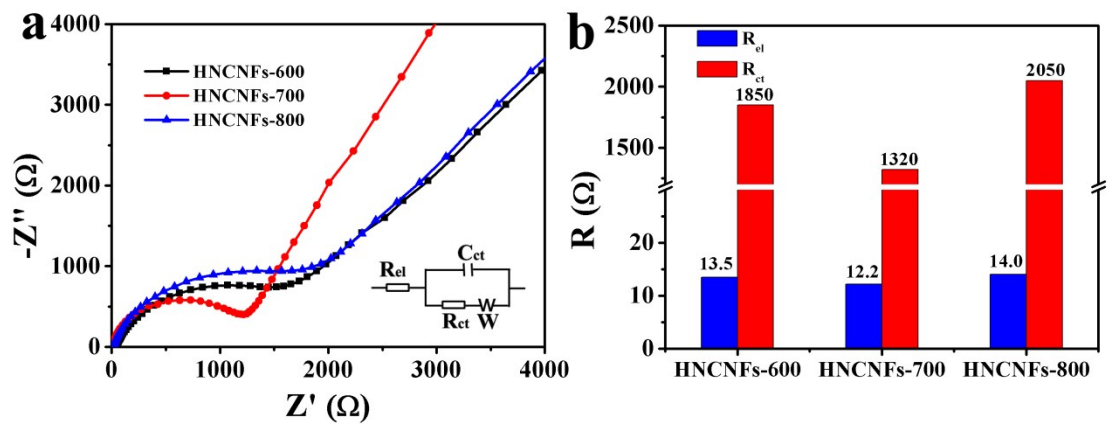


Fig. S11 (a) EIS of HNCNFs-600, HNCNFs-700 and HNCNFs-800 electrodes in fresh PIBs, where the inset shows the corresponding equivalent circuit diagram. (b) The electrolyte resistance (R_{el}) and charge transfer resistance (R_{ct}) values of HNCNFs-600, HNCNFs-700 and HNCNFs-800 electrodes.

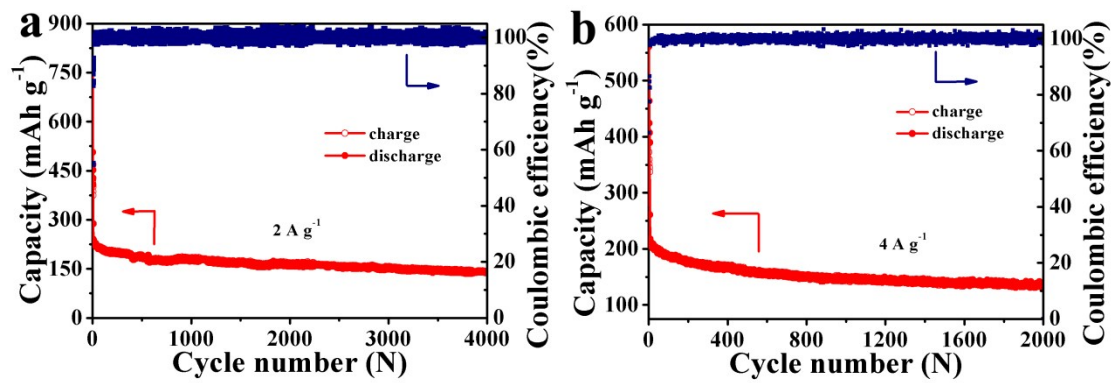


Fig. S12 Cycling performance of the HNCNFs-700 electrode as well as the coulombic efficiency at (a) 2 A g⁻¹ for 4000 cycles and (b) 4 A g⁻¹ for 2000 cycles.

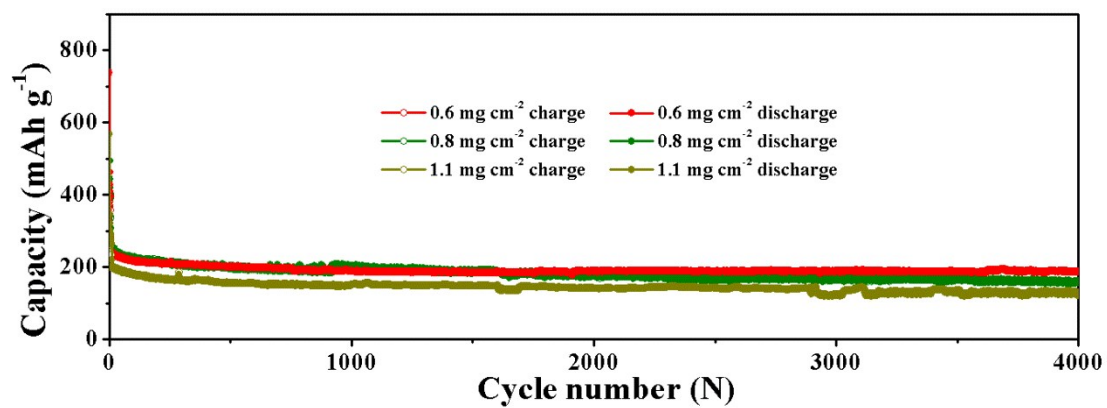


Fig. S13 Cycling performance of the HNCNFs-700 electrode with different mass loadings at 1 A g⁻¹ for 4000 cycles.

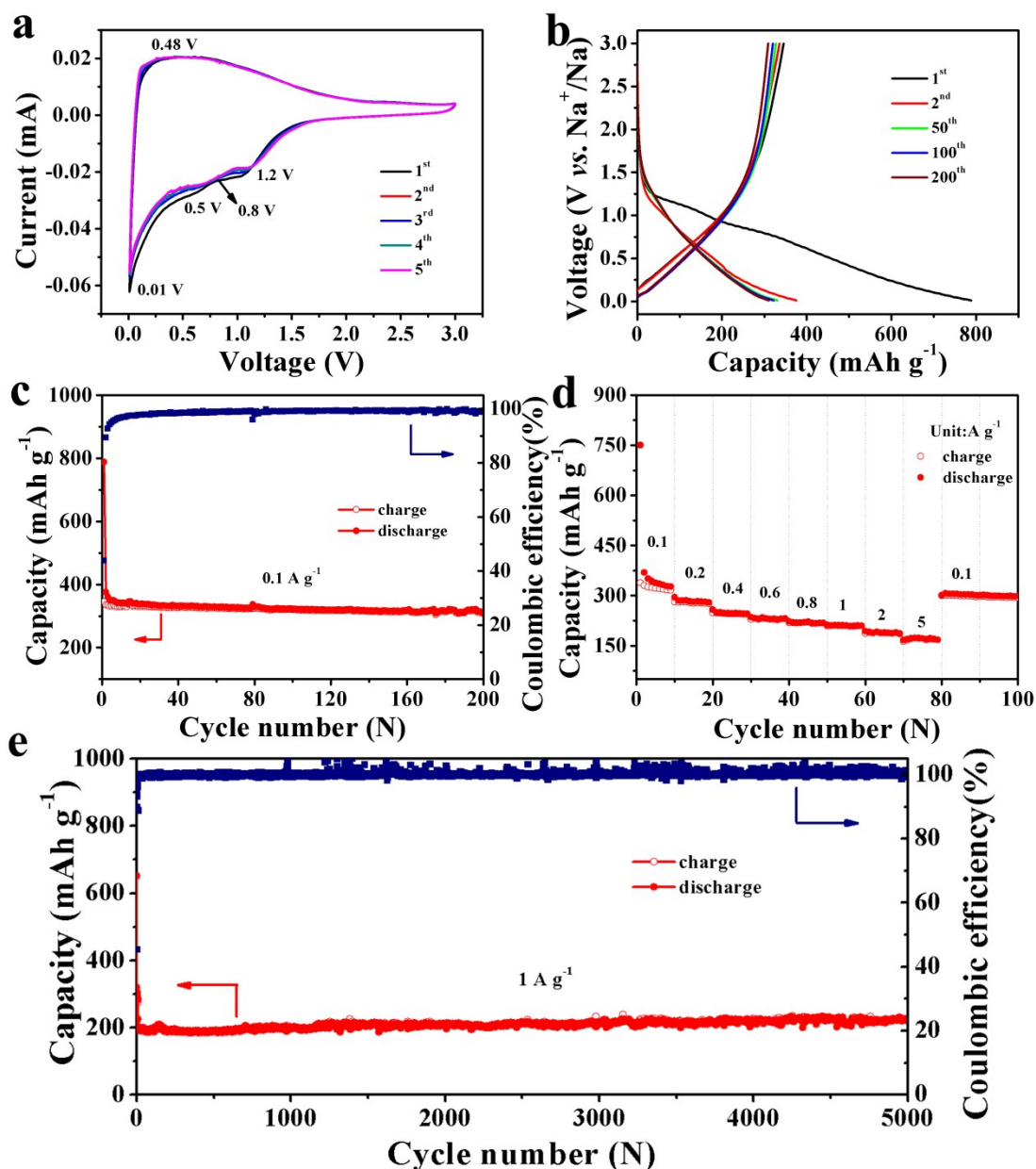


Fig. S14 Electrochemical performances of the HNCNFs-700 electrode in SIBs. (a) CV curves of the HNCNFs-700 electrode at a scan rate of 0.1 mV s⁻¹. (b) Galvanostatic charge/discharge curves of the HNCNFs-700 electrode for the 1st, 2nd, 50th, 100th and 200th cycles at 0.1 A g⁻¹. (c) Cycling performance of the HNCNFs-700 electrode as well as the coulombic efficiency at 0.1 A g⁻¹. (d) Rate performance of the HNCNFs-700 electrode at various current densities. (e) Long cycling performance of the HNCNFs-700 electrode as well as the coulombic efficiency at 1 A g⁻¹.

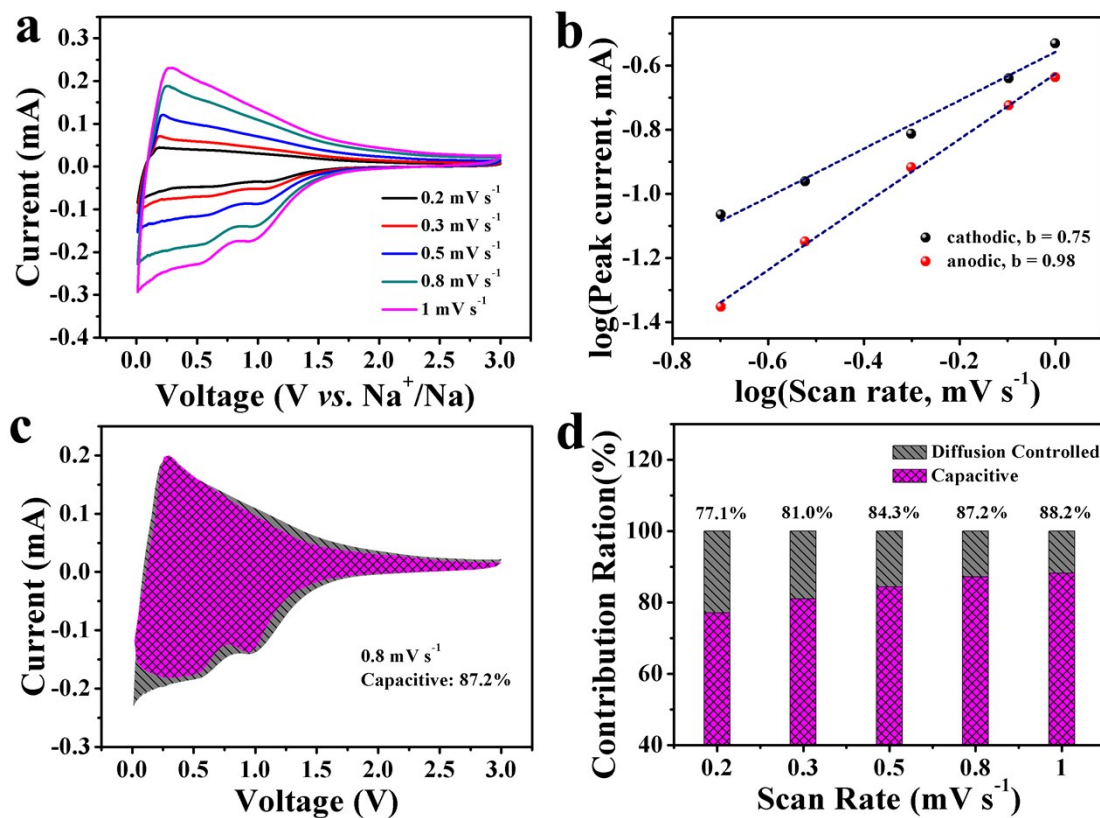


Fig. S15 Electrochemical kinetics analysis in SIBs. (a) CV curves of the HNCNFs-700 electrode at different scan rates from 0.2 to 1 mV s⁻¹. (b) Measurement of b value with the relationship between $\log(i)$ and $\log(v)$. (c) Capacitive and diffusion-controlled contributions to the charge storage at 0.8 mV s⁻¹. (d) Normalized contribution ratios of capacitive and diffusion-controlled capacities of the HNCNFs-700 electrode at different scan rates.

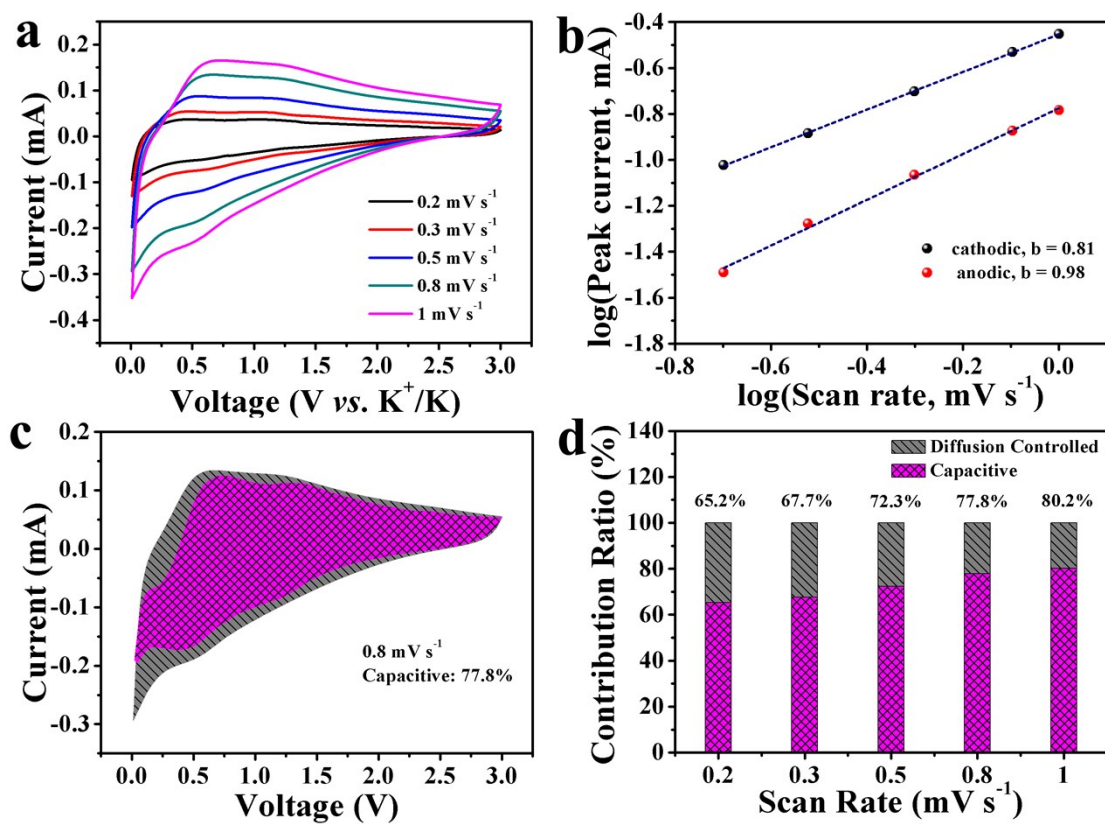


Fig. S16 Electrochemical kinetics analysis. (a) CV curves of the HNCNFs-600 electrode at different scan rates from 0.2 to 1 mV s⁻¹. (b) Measurement of *b* value with the relationship between log(*i*) and log(*v*). (c) Capacitive and diffusion-controlled contributions to the charge storage at 0.8 mV s⁻¹. (d) Normalized contribution ratios of capacitive and diffusion-controlled capacities of the HNCNFs-600 electrode at different scan rates.

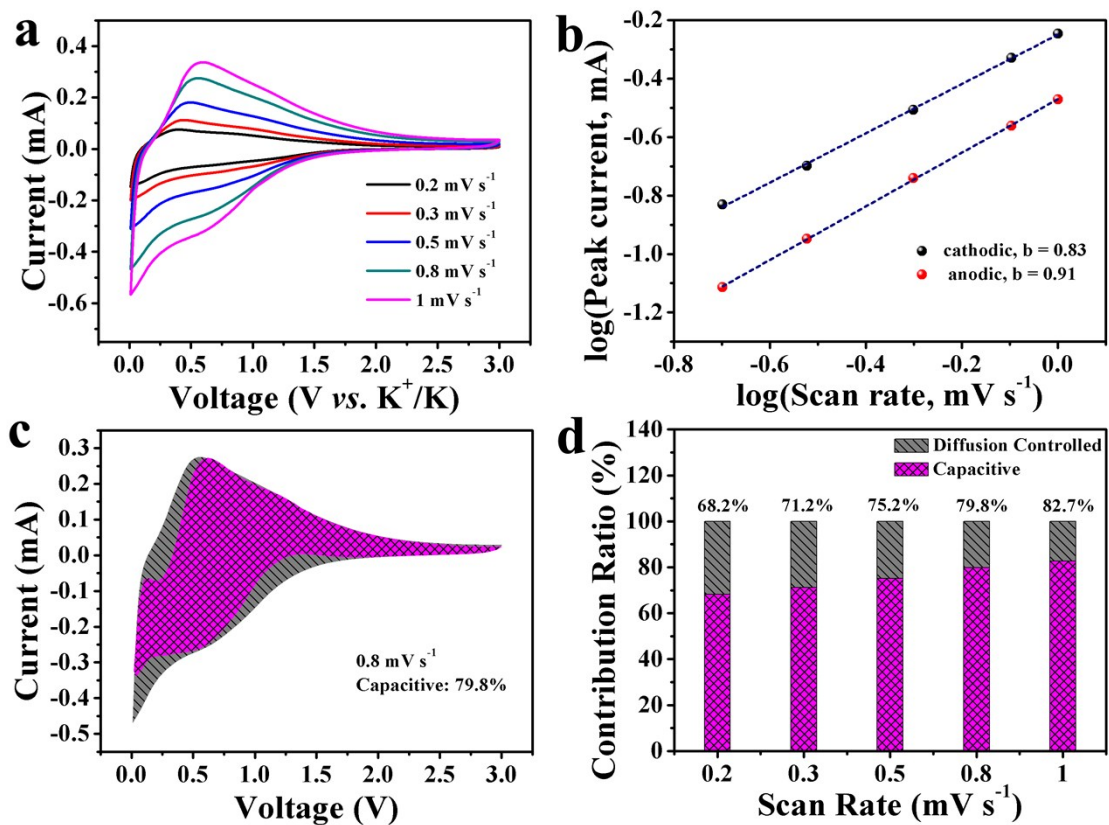


Fig. S17 Electrochemical kinetics analysis. (a) CV curves of the HNCNFs-800 electrode at different scan rates from 0.2 to 1 mV s⁻¹. (b) Measurement of b value with the relationship between $\log(i)$ and $\log(v)$. (c) Capacitive and diffusion-controlled contributions to the charge storage at 0.8 mV s⁻¹. (d) Normalized contribution ratios of capacitive and diffusion-controlled capacities of the HNCNFs-800 electrode at different scan rates.

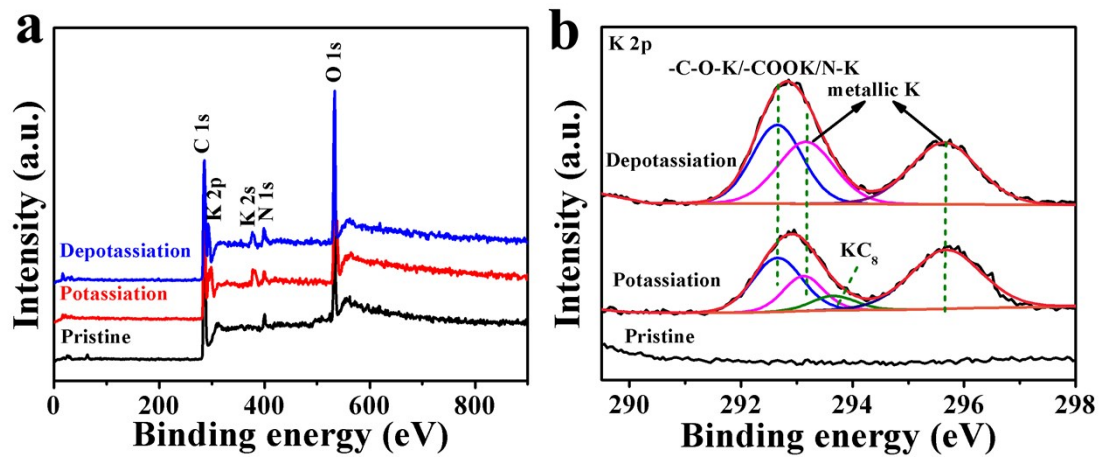


Fig. S18 *Ex-situ* XPS analysis. (a) XPS survey spectra of the HNCNFs-700 electrode under the pristine, potassiation and depotassiation states. (b) High-resolution XPS spectra of K 2p.

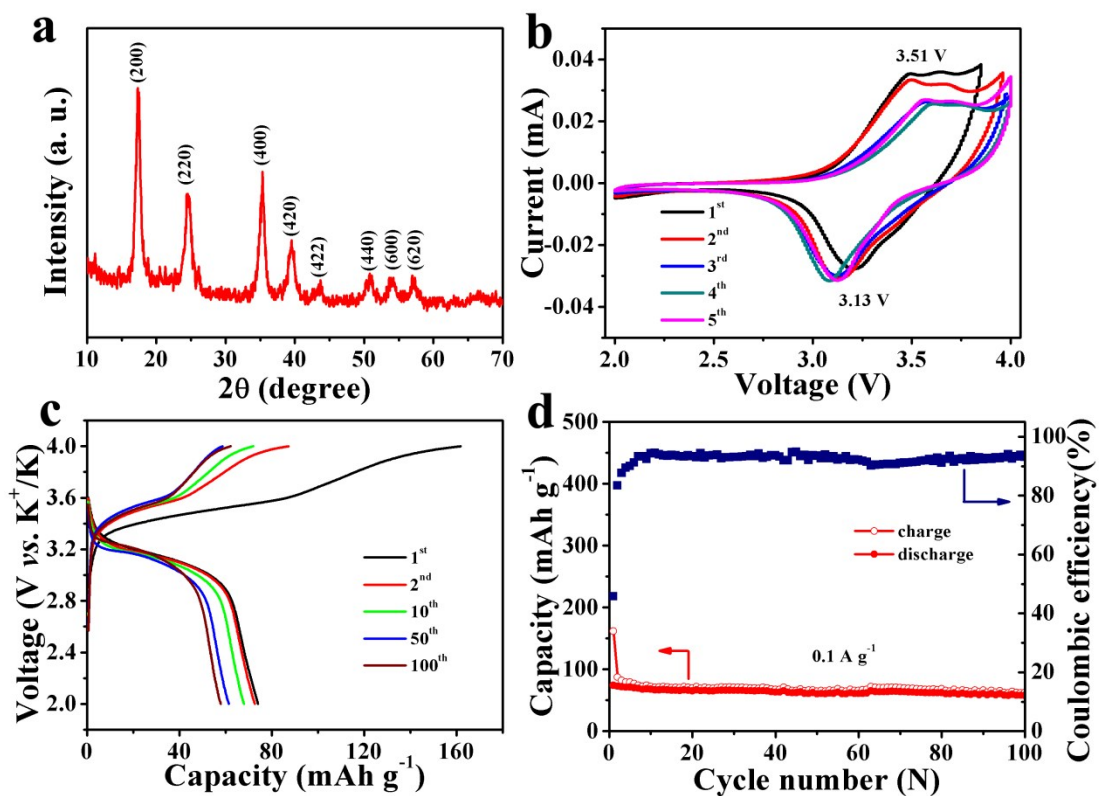


Fig. S19 Structural characterization and electrochemical performance of KPBNPs in a half cell. (a) XRD pattern. (b) CV curves of the KPBNPs electrode at a scan rate of 0.1 mV s^{-1} . (c) Galvanostatic charge/discharge curves of the KPBNPs electrode for the 1st, 2nd, 10th, 50th and 100th cycles at 0.1 A g^{-1} . (d) Cycling performance of the KPBNPs cathode as well as the coulombic efficiency at 0.1 A g^{-1} .

Supplementary Table

Table S1. Structure properties and surface chemistry of three different HNCNFs.

Materials	d_{002} (nm)	I_D/I_G	S_{BET} ($m^2 g^{-1}$)	C (at.%)	N (at.%)	O (at. %)	% of total N 1s		
							pyridinic N	pyrrolic N	graphitic N
HNCNFs-600	0.416	1.67	29.6	84.5	10.3	5.2	31.3	59.8	8.9
HNCNFs-700	0.409	1.59	34.3	85.2	8.2	6.6	38.1	45.7	16.2
HNCNFs-800	0.401	1.42	26.1	88.3	7.4	4.3	31.7	44.9	23.4

Table S2. Electrochemical properties of three different HNCNFs.

Materials	Initial discharge/charge capacity (mAh g ⁻¹)	Initial CE (%)	Capacity (mAh g ⁻¹) ^a	Rate performance (mAh g ⁻¹) ^b	Cycling performance (mAh g ⁻¹) ^c	R_{ct} (Ω)
HNCNFs-600	624.7/312.1	49.9	223.2	121.6	60.5	1850
HNCNFs-700	781.9/410.6	52.5	274.5	139.7	188.4	1320
HNCNFs-800	715.6/374.2	52.3	206.4	70.3	89.5	2050

^a Discharge capacities of the HNCNFs electrodes at 0.1 A g⁻¹ after 200 cycles.

^b Discharge capacities of the HNCNFs electrodes at 30 A g⁻¹.

^c Discharge capacities of the HNCNFs electrodes after 4000 cycles at 1 A g⁻¹.

Table S3. Comparisons of electrochemical properties of some carbon-based electrode materials in PIBs reported in open literatures.

Materials	Initial CE	Rate performance	Cycling performance	Reference
HNCNFs-700	52.5%	239.6 mAh g ⁻¹ (2 A g ⁻¹) 211.0 mAh g ⁻¹ (5 A g ⁻¹) 190.2 mAh g ⁻¹ (10 A g ⁻¹) 161.7 mAh g ⁻¹ (20 A g ⁻¹) 139.7 mAh g ⁻¹ (30 A g ⁻¹)	274.5 mAh g ⁻¹ (200 cycles, 0.1 A g ⁻¹) 188.4 mAh g ⁻¹ (4000 cycles, 1 A g ⁻¹) 141.7 mAh g ⁻¹ (4000 cycles, 2 A g ⁻¹) 132.5 mAh g ⁻¹ (2000 cycles, 4 A g ⁻¹)	This work
Necklace-like N-doped hollow carbon	~30%	224.3 mAh g ⁻¹ (1 A g ⁻¹) 204.8 mAh g ⁻¹ (2 A g ⁻¹)	293.5 mAh g ⁻¹ (400 cycles, 0.2 A g ⁻¹) 161.3 mAh g ⁻¹ (1600 cycles, 1 A g ⁻¹)	Ref. [18] of the text
3D N-doped framework carbon	24.3%	168 mAh g ⁻¹ (1 A g ⁻¹) 146 mAh g ⁻¹ (2 A g ⁻¹) 115 mAh g ⁻¹ (5 A g ⁻¹) 111 mAh g ⁻¹ (10 A g ⁻¹)	137 mAh g ⁻¹ (1000 cycles, 2 A g ⁻¹)	[1]
N/O dual-doped carbon network	47.12%	205 mAh g ⁻¹ (1 A g ⁻¹) 181 mAh g ⁻¹ (2 A g ⁻¹) 175 mAh g ⁻¹ (5 A g ⁻¹)	260 mAh g ⁻¹ (100 cycles, 0.1 A g ⁻¹) 160 mAh g ⁻¹ (4000 cycles, 1 A g ⁻¹)	Ref. [39] of the text
N-doped porous carbon	48.9%	185 mAh g ⁻¹ (10 A g ⁻¹)	384 mAh g ⁻¹ (500 cycles, 0.1 A g ⁻¹) 226.1 mAh g ⁻¹ (1000 cycles, 1 A g ⁻¹) 194 mAh g ⁻¹ (1000 cycles, 2 A g ⁻¹) 160.5 mAh g ⁻¹ (1000 cycles, 5 A g ⁻¹)	Ref. [30] of the text
Highly N-doped carbon fibers	49%	153 mAh g ⁻¹ (2 A g ⁻¹) 126 mAh g ⁻¹ (5 A g ⁻¹) 104 mAh g ⁻¹ (10 A g ⁻¹) 101 mAh g ⁻¹ (20 A g ⁻¹)	248 mAh g ⁻¹ (100 cycles, 0.05 A g ⁻¹) 164 mAh g ⁻¹ (2000 cycles, 1 A g ⁻¹) 146 mAh g ⁻¹ (4000 cycles, 2 A g ⁻¹)	Ref. [19] of the text
N/O dual-doped hierarchical porous hard carbon	25%	118 mAh g ⁻¹ (3 A g ⁻¹)	230.6 mAh g ⁻¹ (100 cycles, 0.05 A g ⁻¹) 130 mAh g ⁻¹ (1100 cycles, 1050 mA g ⁻¹)	Ref. [29] of the text
Amorphous ordered mesoporous carbon	63.6%	144 mAh g ⁻¹ (1 A g ⁻¹)	257.4 mAh g ⁻¹ (100 cycles, 0.05 A g ⁻¹) 146.5 mAh g ⁻¹ (1000 cycles, 1 A g ⁻¹)	Ref. [4] of the text
Ultra-high pyridinic N-doped porous carbon	20%	178 mAh g ⁻¹ (5 A g ⁻¹)	260 mAh g ⁻¹ (120 cycles, 0.05 A g ⁻¹) 152 mAh g ⁻¹ (3000 cycles, 1 A g ⁻¹)	[2]
Free-standing porous carbon nanofibers paper	24.1%	190 mAh g ⁻¹ (2 A g ⁻¹) 140 mAh g ⁻¹ (5 A g ⁻¹) 100 mAh g ⁻¹ (7.7 A g ⁻¹)	270 mAh g ⁻¹ (80 cycles, 0.02 A g ⁻¹) 211 mAh g ⁻¹ (1200 cycles, 0.2 A g ⁻¹)	Ref. [13] of the text

Table S4. The mass loadings of some carbon-based anodes in PIBs reported in open literatures.

Active materials	Mass loading (mg cm ⁻²)	Cycling performance	Reference
HNCNFs-700	0.6	188.4 mAh g ⁻¹ (1 A g ⁻¹ , 4000 cycles)	This work
		141.7 mAh g ⁻¹ (2 A g ⁻¹ , 4000 cycles)	
		132.5 mAh g ⁻¹ (4 A g ⁻¹ , 4000 cycles)	
HNCNFs-700	0.8	161.2 mAh g ⁻¹ (1 A g ⁻¹ , 4000 cycles)	This work
HNCNFs-700	1.1	130.4 mAh g ⁻¹ (1 A g ⁻¹ , 4000 cycles)	This work
N-doped soft carbon frameworks	1.0	165 mAh g ⁻¹ (1 A g ⁻¹ , 500 cycles)	[3]
N/O dual-doped carbon network	0.8	160 mAh g ⁻¹ (1 A g ⁻¹ , 4000 cycles)	Ref. [44] of the text
N-doped carbon nanotubes	1.2	60 mAh g ⁻¹ (2 A g ⁻¹ , 1000 cycles)	[4]
N-doped framework carbon	0.7-1.0	137 mAh g ⁻¹ (2 A g ⁻¹ , 1000 cycles)	[5]
soft Carbon	1.0-1.5	200 mAh g ⁻¹ (20 mA g ⁻¹ , 100 cycles)	[6]
N/S codoped carbon microboxes	1.2	180 mAh g ⁻¹ (0.5 A g ⁻¹ , 1000 cycles)	Ref. [1] of the text
porous thin carbon shells	1.0-1.2	65 mAh g ⁻¹ (2 A g ⁻¹ , 900 cycles)	[7]
graphite foam	1.9	200 mAh g ⁻¹ (40 mA g ⁻¹ , 200 cycles)	[8]
carbon nanotubes-interweaved layer on graphite flakes	0.8	234.4 m Ah g ⁻¹ (2 A g ⁻¹ , 1500 cycles)	[9]
N/S co-doped porous carbon	1.2	125 mAh g ⁻¹ (1 A g ⁻¹ , 1000 cycles)	[10]
N-doped porous carbon	0.64	226.1 mAh g ⁻¹ (1 A g ⁻¹ , 1000 cycles)	Ref. [30] of the text
N-doped bamboo-like carbon nanotubes	0.5	204 mAh g ⁻¹ (0.5 A g ⁻¹ , 1000 cycles)	[11]
hollow carbon nanospheres	0.2	144.3 mAh g ⁻¹ (4 A g ⁻¹ , 2000 cycles)	Ref. [11] of the text

graphitic carbon nanocage	0.8-1.0	248 mAh g ⁻¹ (55 mA g ⁻¹ , 100 cycles)	Ref. [49] of the text
N-doped carbon nanotubes	0.7	102 mAh g ⁻¹ (2 A g ⁻¹ , 500 cycles)	[12]
few layer N-doped graphene	0.63-0.75	150 mAh g ⁻¹ (0.5 A g ⁻¹ , 500 cycles)	[13]
high pyridine N-doped porous carbon	0.8	231 mAh g ⁻¹ (0.5 A g ⁻¹ , 2000 cycles)	Ref. [15] of the text
amorphous ordered mesoporous carbon	1.0	146 mAh g ⁻¹ (1 A g ⁻¹ , 1000 cycles)	Ref. [4] of the text
highly N-doped carbon nanofibers	1.5	164 mAh g ⁻¹ (1 A g ⁻¹ , 2000 cycles)	Ref. [19] of the text
N-doped carbon nanofibers	0.9-1.3	103.4 mAh g ⁻¹ (0.5 A g ⁻¹ , 1000 cycles)	Ref. [41] of the text
expanded graphite	2.0	174 mAh g ⁻¹ (0.2 A g ⁻¹ , 500 cycles)	Ref. [43] of the text
highly disordered hard carbon	1.0	240 mAh g ⁻¹ (0.2 A g ⁻¹ , 150 cycles)	[14]
3D rGO aerogel	0.7-1.0	137 mAh g ⁻¹ (2 A g ⁻¹ , 1000 cycles)	[15]
N/O dual-doped hard carbon	0.9	130 mAh g ⁻¹ (1050 mA g ⁻¹ , 1100 cycles)	Ref. [29] of the text
hard-soft composite carbon	2.0	200 mAh g ⁻¹ (0.2 A g ⁻¹ , 200 cycles)	[16]
ultra-high pyridinic N-doped porous carbon	1.0	152 mAh ⁻¹ (1 A g ⁻¹ , 3000 cycles)	[2]
few-layer F-doped graphene foam	0.52-0.58	165.9 mAh g ⁻¹ (0.5 A g ⁻¹ , 200 cycles)	[17]

Table S5. Energy density (E) of carbon-based potassium-ion full cells reported in open literatures.

Materials	E (Wh kg ⁻¹)	Reference
HNCNFs-700//KPBNPs	70.3	This work
carbon foam//carbon foam	58	[18]
N-doped carbon nanotubes//laser scribed graphene	65	[4]
activated carbon//K ₂ TP	101	[19]
onion-like carbon//activated carbon	142	[20]
N, P-codoped graphene grown on carbon cloth//KPB	231.5	[21]
graphenic carbon//K ₂ Ti ₆ O ₁₃	58.2	[22]
activated carbon//Ca _{0.5} Ti ₂ (PO ₄) ₃ @C	80	[23]
graphite//prussian blue	110	Ref. [55] of the text
soft carbon//commercialized activated carbon	120	[24]
N-doped porous carbon//PTCDA	153	[2]

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